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2. Patent Application number (The Patent Office will fill in this part)	02 AUG 2002 0217990.1		
3. Full name, address and postcode of the or each applicant (underline all surnames)	IMPERIAL COLLEGE INNOVATIONS LIMITED Sherfield Building Exhibition Road London SW7 2AZ		
Patents ADP Number (if you know it)	07409436002		
If the applicant is a corporate body, give the country/state of its incorporation	United Kingdom		
4. Title of the invention	Low-Temperature Metal Oxide Coating		
5. Name of your agent (if you have one)	FRY HEATH & SPENCE		
"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)	The Old College 53 High Street Horley, Surrey, RH6 7BN		
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Description 11

Claim(s) 5

Abstract 1

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# DUPLICATE

## LOW-TEMPERATURE METAL OXIDE COATING

The present invention relates to a low-temperature deposition method for the deposition of metal oxide coatings, in particular the conformal coating of surfaces, especially structured surfaces, flat surfaces and particles. In particular, the low-temperature deposition method is suited to the conformal coating of reticulated films, nanocrystalline films and nanoparticles.

Several methods have been disclosed for the deposition of metal oxide coatings.

One such method, and the most common method, is based on precursor pyrolysis. Typically, in this method, a colloidal suspension of semiconductor particles to be coated, for example,  $\text{SnO}_2$ ,  $\text{TiO}_2$  or  $\text{ZnO}$  particles, is prepared in a solution containing a precursor of the desired coating oxide, for example, aluminium acetate for an  $\text{Al}_2\text{O}_3$  coating [1]. The coated particles are then subsequently subjected to a high-temperature heat treatment at a temperature of at least  $150^\circ\text{C}$ , and typically about  $450^\circ\text{C}$ , in order to provide for a complete reaction of the precursor to the oxide.

In another such method, a reticulated film is coated with a chloride precursor, for example,  $\text{AlCl}_3$  or  $\text{MgCl}_2$  diluted in an alcoholic solution. The coated film is then subjected to a high-temperature heat treatment in order to provide for a complete reaction of the precursor to the oxide.

In a further such method, templates are utilized. The templates are typically surfactant micelles [3], such as cetyltrimethylammonium chloride. Following coating, the coated substrate is typically subjected to a heat treatment at a temperature of  $100^\circ\text{C}$  for 48 hours in an enclosed reactor, and subsequently, in order to remove the templates, a high-temperature heat treatment at a temperature of  $450^\circ\text{C}$  for 2 hours.

These methods each require a high-temperature heat treatment, and thus, in particular, are not suited to the coating of temperature-sensitive substrates. The requirement for a high-temperature heat treatment is a significant limitation to the applicability of these

methods, in limiting the techniques to temperature-stable substrates, such as glass, and the coatings to ones which comprise temperature-stable components.

A low-temperature deposition method has been developed for the deposition of  
5 amorphous metal oxides onto substrates, as embodied a gold-coated quartz crystal  
microbalance (QCM) [4]. In this method, a precursor solution is prepared of a metal  
alkoxide precursor in an organic solvent, for example, toluene and ethanol. The metal  
alkoxide in the precursor solution is partially hydrolysed, resulting in a sol. The substrate  
is then coated by dipping the substrate in the sol, and subsequently rinsed with water. In  
10 this method, the formation of a sol is required prior to coating. As the sol comprises  
partially-polymerised metal oxide suspended in solution, which is an optical scattering  
solution, the method is particularly unsuited to the conformal coating of reticulated or  
nanocrystalline films.

15 It is an aim of the present invention to provide a low-temperature deposition method for  
the deposition of metal oxide coatings, in particular the conformal coating of reticulated  
films, nanocrystalline films and nanoparticles.

In one aspect the present invention provides a low-temperature deposition method for  
20 depositing metal oxide coatings on a substrate, the method comprising the steps of:  
coating a surface of a substrate with a non-hydrolysed precursor solution of one or more  
moisture-sensitive metal alkoxides in an organic solvent at a temperature of less than 150  
°C; and hydrolysing the precursor solution coated on the surface of the substrate to form  
a metal oxide coating at a temperature of less than 150 °C.

25

Preferably, the one or more moisture-sensitive alkoxides comprise  $M(OR)_z$ , where M is  
any metal, and OR is an alkoxide group.

More preferably, the metal is a metal selected from the group consisting of Al, Ce, Mg,  
30 Nb, Si, Sn, Ti, Va, Zn and Zr.

Preferably, the step of coating a surface of a substrate is performed at room temperature.

In one embodiment the step of coating a surface of a substrate comprises the step of dipping the surface of the substrate in the precursor solution.

Preferably, the surface of the substrate is dipped in the precursor solution for a period of up to about 1 hour, more preferably from about 1 minute to about 1 hour, and even more preferably from about 10 minutes to about 1 hour.

In another embodiment the step of coating a surface of a substrate comprises the step of spraying the surface of the substrate with the precursor solution.

10

In a further embodiment the step of coating a surface of a substrate comprises the step of spin-coating the surface of the substrate with the precursor solution.

Preferably, the step of hydrolysing the precursor solution coated on the surface of the substrate is performed at room temperature.

15

Preferably, the step of hydrolysing the precursor solution coated on the surface of the substrate comprises the step of rinsing the surface of the substrate coated in the precursor solution with water.

20

Preferably, the coating is a conformal coating.

In one embodiment the surface of the substrate comprises a flat surface.

25 In one embodiment the surface of the substrate comprises a nanocrystalline film.

In another embodiment the surface of the substrate comprises a particulate solid film.

In a further embodiment the surface of the substrate comprises a structured surface.

30

Preferably, the structured surface comprises a reticulated film.

In one embodiment the substrate includes a temperature-sensitive element.

In one embodiment the temperature-sensitive element is selected from the group consisting of a plastic and a polymer.

- 5 In another embodiment the temperature-sensitive element comprises temperature-sensitive molecules.

In one preferred embodiment the molecules are selected from the group consisting of inorganic, organic and organometallic molecules.

10

In another preferred embodiment the molecules are polymers.

In a further preferred embodiment the molecules are biomolecules.

- 15 In a yet further preferred embodiment the molecules are biological macromolecules.

Preferably, the biological macromolecules are selected from the group consisting of proteins and nucleic acids.

- 20 In one embodiment the molecules are at the surface of the substrate.

In one embodiment the coating extends over regions of the surface of the substrate not encompassed by the molecules.

- 25 In another embodiment the coating encapsulates the molecules.

In still another embodiment the substrate comprises particles.

In one embodiment the particles comprise dry particles.

30

In another embodiment the particles are suspended in solution.

Preferably, the particles comprise nanoparticles.

In another aspect the present invention provides a non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent.

- 5 Preferably, the one or more moisture-sensitive alkoxides comprise  $M(OR)_x$ , where M is any metal, and OR is an alkoxide group.

More preferably, the metal is a metal selected from the group consisting of Al, Ce, Mg, Nb, Si, Sn, Ti, V, Zn and Zr.

10

In a further aspect the present invention provides a method of preparing a non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent, the method comprising the step of mixing one or more moisture-sensitive metal alkoxides in an organic solvent in a controlled environment containing

- 15 less than about 10 ppm water.

Preferably, the method is performed at room temperature.

Preferably, the controlled environment is an inert atmosphere.

20

Preferably, the one or more moisture-sensitive alkoxides comprise  $M(OR)_x$ , where M is any metal, and OR is an alkoxide group.

More preferably, the metal is a metal selected from the group consisting of Al, Ce, Mg,

- 25 Nb, Si, Sn, Ti, V, Zn and Zr.

The low-temperature coating method of the present invention allows for the deposition of coatings having a thickness of less than one nanometer to hundreds of nanometers, with repeated deposition allowing for the deposition of coatings of increased thickness.

30

In the context of the present invention, low temperature relates to temperatures of less than 150 °C, in particular encompassing the deposition of coatings at room temperature.



Low-temperature processing is attractive in reducing cost and environmental waste, and, moreover, allows for the coating of temperature-sensitive substrates, in particular organic substrates, such as polymers and plastics.

5 The present invention finds particular application in the conformal coating of highly-reticulated inorganic films. Such films are utilized in a wide range of photochemical, photocatalytic, optoelectronic and electronic devices. Particular examples are the use of mesoporous, nanocrystalline metal oxide films for optoelectronic devices, such as photovoltaic or photoelectrochemical solar cells, light-emitting devices, and  
10 photocatalytic devices for the decomposition of pollutants or the photocatalytic scavenging of oxygen from closed environments.

The conformal coating of reticulated inorganic films with a thin layer, typically from about 0.2 nm to about 10 nm, of a metal oxide is particularly attractive in enabling  
15 control of the surface properties of such films. For example, the fabrication of conformal insulating layers on nanocrystalline metal oxide films provides for the retardation of interfacial recombination processes. Such barrier layers could comprise low-electron affinity metal oxides, such as  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{SiO}_2$  or  $\text{ZrO}_2$ . These coatings would be particularly attractive for device applications including photovoltaic cells and  
20 photochromic films.

Another application is the coating of metal oxide particles with a barrier layer to prevent photocatalytic, photochemical or other activity. Such passivated particles, for example,  $\text{TiO}_2$  particles coated with  $\text{Al}_2\text{O}_3$ , are widely used as whiteners or light scatterers in the  
25 pigment, dye and cosmetic industries.

A further application is the coating of flat substrates, including temperature-sensitive substrates, such as plastics, for example, in order to provide an electrically-insulating barrier layer. Such substrates include ITO or  $\text{F:SnO}_2$  coated plastic or glass.

30

A yet further application is the coating of metal oxide films, in particular reticulated and nanocrystalline films, having temperature-sensitive molecules pre-absorbed thereon. Such molecules include inorganic, organic and organometallic molecules, polymers,

biomolecules and biological macromolecules, such as proteins and nucleic acids. In one embodiment the coating can extend over regions of the surface of the substrate which are not encompassed by the temperature-sensitive molecules. In another embodiment the coating can encapsulate the temperature-sensitive molecules.

5

Preferred embodiments of the present invention will now be described hereinbelow by way of example only with reference to the accompanying drawings, in which:

Figure 1 illustrates high-resolution TEMs of (a, b) an  $\text{Al}_2\text{O}_3$  overlayer coated on a nanocrystalline  $\text{TiO}_2$  film, and (c, d) an uncoated nanocrystalline  $\text{TiO}_2$  film; and

10

Figure 2 illustrates the current-voltage characteristics of a dye-sensitized, nanocrystalline sandwich solar cell structure comprising (a) a  $\text{TiO}_2$  film having an  $\text{Al}_2\text{O}_3$  conformal coating, and (b) an uncoated  $\text{TiO}_2$  film.

15

In a first step, a stable, non-hydrolysed precursor solution is prepared of one or more moisture-sensitive metal alkoxides in an organic solvent.

The moisture-sensitive alkoxides can be expressed generally as  $\text{M}(\text{OR})_z$ , where M is any metal, OR is an alkoxide group and z is the valence or oxidation state of the metal. In preferred embodiments the metal is a metal selected from the group consisting of Al, Ce, Mg, Nb, Si, Sn, Ti, Va, Zn and Zr.

20

The metal alkoxide precursors are diluted in an organic solvent at room temperature, typically at about 25 °C. Examples of such solutions include a 0.15 M solution of aluminum tri-sec-butoxide in dry iso-propanol, a 0.15 M solution of silicon methoxide in dry methanol, and a 0.15 M of zirconium iso-butoxide in dry iso-propanol.

25

In a preferred embodiment the precursor solution is prepared in an inert atmosphere, here nitrogen, and under a strictly-controlled water presence, here less than about 10 ppm, in order to avoid partial sol-gel hydrolysis. In this embodiment the controlled environment for the precursor solution is provided in a glove box.

30

The present inventors have recognized that, with suitable solvent and precursor concentrations, the precursor solution is rendered insensitive to the atmosphere. The precursor solution is stable for several months under normal atmosphere; the solution remaining clear without any visible precursor hydrolysis.

5

In a second, coating step, a substrate is coated with the precursor solution, in this embodiment by dipping the substrate in the precursor solution for a period of from about 10 minutes to 1 hour at room temperature under aerobic conditions. In alternative embodiments the substrate can be coated using alternative coating techniques, such as spraying or spin-coating.

10

In a third, rinsing step, the coated surface of the substrate is rinsed with water.

15

Where thicker films are required, the coating procedure of the coating and rinsing steps is repeated as required.

In the coating step, the non-hydrolysed metal alkoxides start reacting with the hydroxylated surface of the substrate, leading to the formation of a primary shell.

20

In the rinsing step, the rinsing of the surface of the substrate with water drives to completion the hydrolysis of the metal alkoxides, and causes the formation of intrapolymeric branches and bonds between the primary metal alkoxides adsorbed on the surface of the substrate. As the metal alkoxide precursors are moisture sensitive, the precursors are fully hydrolysed in the presence of water, allowing the reaction to go to completion during the rinsing step, thereby obviating the requirement for a high-temperature heat treatment step. Following the rinsing step, a hydroxylated surface is formed on the new metal oxide shell, which hydroxylated surface enables further coating, if desired, and thereby a homogeneous increase in the shell thickness.

25

A particular advantage of the coating method of the present invention is in enabling the deposition of conformal metal oxide coatings on reticulated or particulate substrates, without the requirement for any high-temperature heat treatment, that is, a heat treatment at a temperature typically above 150 °C.

30

The present invention will now be described hereinbelow by way of example only with reference to the following non-limiting Example.

5 Example

This Example is directed to the fabrication of a nanocrystalline sandwich solar cell structure comprising an  $\text{RuL}_2(\text{NCS})_2$  sensitized  $\text{Al}_2\text{O}_3$  conformal coating on a  $\text{TiO}_2$  film.

10 In a first, precursor preparation step, a 0.15 M precursor solution was prepared of aluminum tri-sec-butoxide in dry iso-propanol.

In a second, coating step, a preformed mesoporous, nanocrystalline  $\text{TiO}_2$  film having a thickness of 8  $\mu\text{m}$  was coated with the precursor solution by dipping the film in the  
15 precursor solution at room temperature for 10 minutes. Such  $\text{TiO}_2$  films can be fabricated either by sol-gel chemistry on glass substrates, or by high-pressure compression of nanoparticles on plastic substrates.

The coated film was then rinsed in de-ionized water to form an  $\text{Al}_2\text{O}_3$  conformal coating.  
20

The  $\text{Al}_2\text{O}_3$  conformal coating was dried in nitrogen gas, and then sensitized overnight in a 1 mM solution of  $\text{RuL}_2(\text{NCS})_2$  in 1:1 acetonitrile/tert-butanol.

Figures 1(a) and (b) illustrate high-resolution TEMs of a fragment of the deposited  $\text{Al}_2\text{O}_3$   
25 conformal coating, with Figure 1(b) being at higher magnification. For reference, Figures 1(c) and (d) illustrate high-resolution TEMs of a fragment of an uncoated nanocrystalline  $\text{TiO}_2$  film, with Figures 1(c) and (d) being at the same respective magnifications as Figures 1(a) and (b). The high-resolution TEM data shows the thickness of the  $\text{Al}_2\text{O}_3$  conformal coating at about 1 nm.

30

Figure 2(a) illustrates the current-voltage characteristics of the resulting dye-sensitized structure. For reference, Figure 2(b) illustrates the current-voltage characteristics of an uncoated nanocrystalline  $\text{TiO}_2$  film. The determined data was obtained for transparent

counter electrodes and a cell active area of  $0.8 \text{ cm}^2$  under AM1.5 simulated sunlight at  $100 \text{ mWcm}^{-2}$ , with the insert showing the corresponding dark current data.

As will be noted, the  $\text{Al}_2\text{O}_3$  conformal coating of the present invention provides a  
5 significant improvement in device performance, with the solar-to-electrical power conversion efficiency increasing by 30 %.

Finally, it will be understood that the present invention has been described in its preferred embodiments and can be modified in many different ways without departing  
10 from the scope of the invention as defined by the appended claims.

Numerous minor modifications of the described coating method can be envisaged to optimize the method for different applications, including minor, low-temperature heat treatments, and different precursors, precursor concentrations and solvents.

## REFERENCES

- [1] Tennakone, K., Perera, V. P. S., Kottegoda, I. R. M., Kumara, G. R. R. A., *J. Phys. D: Appl. Phys.* 1999, 32, pp 374-379.
- 5 [2] Tennakone, K., Bandaranayake, P. K. M., Jayaweera, P. V. V., Kommo, A., Kumara, G. R. R., A., *Physica E* 2002, 14, pp 190-196.
- [3] Elder, S. H., Cot, F. M., Su, Y., Heald, S. M., Tyryshkin, A. M., Bowman, M. K., Gao, Y., Joly, A. G., Balmer, M. L., Kolwaite, A. C., Magrini, K. A., Blake, D. M., *J. Am. Chem. Soc.* 2000, 122, pp 5138-5146.
- 10 [4] Ichinose, I., Senzu, H., Kunitake, T., *Chem. Mater.* 1997, 9, pp 1296-1298.

CLAIMS

1. A low-temperature deposition method for depositing metal oxide coatings on a substrate, the method comprising the steps of:  
5 coating a surface of a substrate with a non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent at a temperature of less than 150 °C; and  
hydrolysing the precursor solution coated on the surface of the substrate to form a metal oxide coating at a temperature of less than 150 °C.  
10
2. The method of claim 1, wherein the one or more moisture-sensitive alkoxides comprise  $M(OR)_x$ , where M is any metal, and OR is an alkoxide group.
3. The method of claim 2, wherein the metal is a metal selected from the group  
15 consisting of Al, Ce, Mg, Nb, Si, Sn, Ti, Va, Zn and Zr.
4. The method of any of claims 1 to 3, wherein the step of coating a surface of a substrate is performed at room temperature.
- 20 5. The method of any of claims 1 to 4, wherein the step of coating a surface of a substrate comprises the step of:  
dipping the surface of the substrate in the precursor solution.
6. The method of claim 5, wherein the surface of the substrate is dipped in the  
25 precursor solution for a period of from about 1 minute to about 1 hour.
7. The method of any of claims 1 to 4, wherein the step of coating a surface of a substrate comprises the step of:  
spraying the surface of the substrate with the precursor solution.  
30
8. The method of any of claims 1 to 4, wherein the step of coating a surface of a substrate comprises the step of:  
spin-coating the surface of the substrate with the precursor solution.

9. The method of any of claims 1 to 8, wherein the step of hydrolysing the precursor solution coated on the surface of the substrate is performed at room temperature.
- 5 10. The method of any of claims 1 to 9, wherein the step of hydrolysing the precursor solution coated on the surface of the substrate comprises the step of:  
rinsing the surface of the substrate coated in the precursor solution with water.
11. The method of any of claims 1 to 10, wherein the coating is a conformal coating.
- 10 12. The method of any of claims 1 to 11, wherein the surface of the substrate comprises a flat surface.
13. The method of any of claims 1 to 12, wherein the surface of the substrate  
15 comprises a nanocrystalline film.
14. The method of any of claims 1 to 12, wherein the surface of the substrate comprises a particulate solid film.
- 20 15. The method of any of claims 1 to 11, wherein the surface of the substrate comprises a structured surface.
16. The method of claim 15, wherein the structured surface comprises a reticulated film.
- 25 17. The method of any of claims 1 to 16, wherein the substrate includes a temperature-sensitive element.
18. The method of claim 17, wherein the temperature-sensitive element is selected  
30 from the group consisting of a plastic and a polymer.
19. The method of claim 17, wherein the temperature-sensitive element comprises temperature-sensitive molecules.



20. The method of claim 19, wherein the molecules are selected from the group consisting of inorganic, organic and organometallic molecules.
- 5 21. The method of claim 19, wherein the molecules are polymers.
22. The method of claim 19, wherein the molecules are biomolecules.
23. The method of claim 19, wherein the molecules are biological macromolecules.
- 10 24. The method of claim 23, wherein the biological macromolecules are selected from the group consisting of proteins and nucleic acids.
25. The method of any of claims 19 to 24, wherein the molecules are at the surface of  
15 the substrate.
26. The method of claim 25, wherein the coating extends over regions of the surface of the substrate not encompassed by the molecules.
- 20 27. The method of claim 25, wherein the coating encapsulates the molecules.
28. The method of any of claims 1 to 11, wherein the substrate comprises particles.
29. The method of claim 28, wherein the particles comprise dry particles.
- 25 30. The method of claim 28, wherein the particles are suspended in solution.
31. The method of any of claims 28 to 30, wherein the particles comprise nanoparticles.
- 30 32. A non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent.

33. The precursor solution of claim 32, wherein the one or more moisture-sensitive alkoxides comprise  $M(OR)_z$ , where M is any metal, and OR is an alkoxide group.

34. The precursor solution of claim 33, wherein the metal is a metal selected from the group consisting of Al, Ce, Mg, Nb, Si, Sn, Ti, Va, Zn and Zr.

35. A method of preparing a non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent, the method comprising the step of mixing one or more moisture-sensitive metal alkoxides in an organic solvent in a controlled environment containing less than about 10 ppm water.

36. The method of claim 35, where performed at room temperature.

37. The method of claim 35 or 36, wherein the controlled environment is an inert atmosphere.

38. The method of any of claims 35 to 37, wherein the one or more moisture-sensitive alkoxides comprise  $M(OR)_z$ , where M is any metal, and OR is an alkoxide group.

39. The method of claim 38, wherein the metal is a metal selected from the group consisting of Al, Ce, Mg, Nb, Si, Sn, Ti, Va, Zn and Zr.

40. A low-temperature deposition method for depositing metal oxide coatings on a substrate substantially as hereinbefore described with reference to Figures 1(a) and (b) and Figure 2(a) of the accompanying drawings.

41. A low-temperature deposition method for depositing metal oxide coatings on a substrate substantially as hereinbefore described with reference to the Example.

42. A non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent substantially as hereinbefore described with reference to Figures 1(a) and (b) and Figure 2(a) of the accompanying drawings.

43. A non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent substantially as hereinbefore described with reference to the Example.

5

44. A method of preparing a non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent substantially as hereinbefore described with reference to Figures 1(a) and (b) and Figure 2(a) of the accompanying drawings.

10

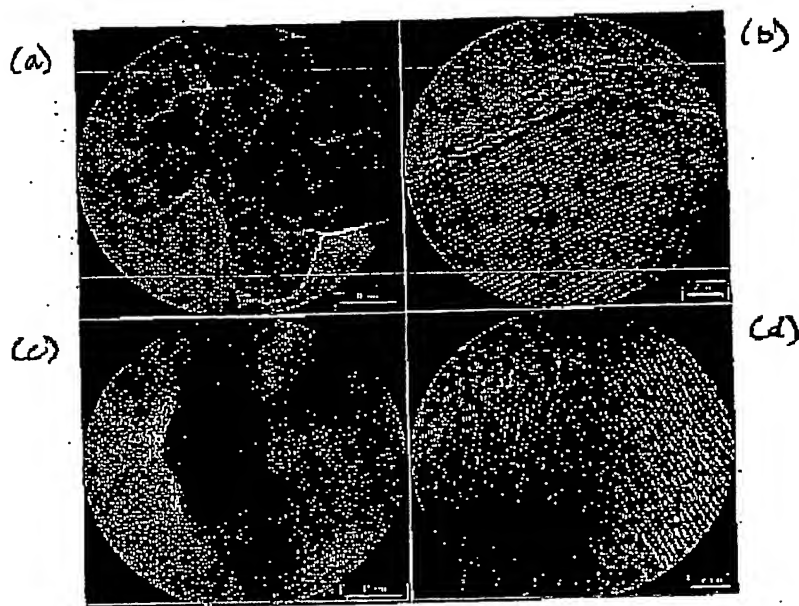
45. A method of preparing a non-hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an organic solvent substantially as hereinbefore described with reference to the Example.

**ABSTRACT****LOW-TEMPERATURE METAL OXIDE COATING**

- 5 A low-temperature deposition method for depositing metal oxide coatings on a substrate,  
the method comprising the steps of: coating a surface of a substrate with a non-  
hydrolysed precursor solution of one or more moisture-sensitive metal alkoxides in an  
organic solvent at a temperature of less than 150 °C; and hydrolysing the precursor  
solution coated on the surface of the substrate to form a metal oxide coating at a  
10 temperature of less than 150 °C.

[Figure 2]

1/2

**Figure 1**

2/2

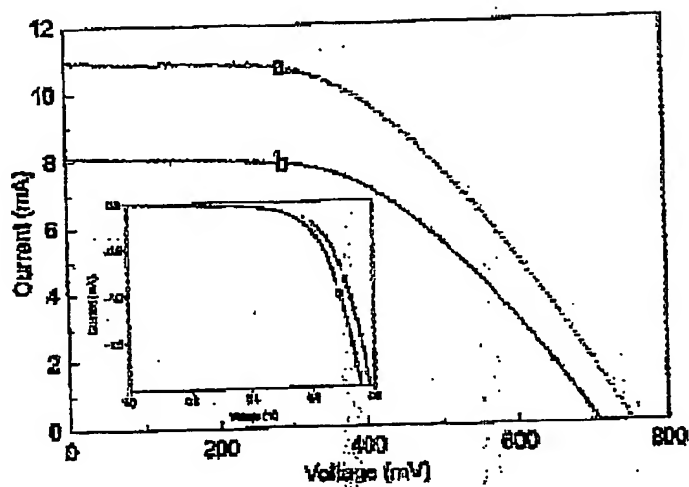


Figure 2

PCT Application

**GB0303397**

